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RADIOCHEMICAL ANALYSES FOR PREOPERATIONAL SURVEILLANCE

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RADIOCHEMICAL ANALYSES FOR PREOPERATIONAL SURVEILLANCE

In the present state of the art in nuclear operations, environmental monitoring is as fundamental as film badges, neutrons, or uranium. Presumably there will come a time when we are sufficiently confident of our ability to design a reactor, a chemical processing plant, or other nuclear facility, that the question of environmental contamination will never arise; that time has not yet come. Preoperational monitoring to date has chiefly consisted of measurements of gross activity; it is the purpose of this paper to show the usefulness of analysis for specific nuclides in this type of measurement.

To the management of a nuclear facility, the primary purposes of environmental monitoring are to establish that no harm has been done to anyone by its operations, and to limit the area of liability if radioactive materials are accidentally discharged in damaging quantities. A subsidiary function is to establish good public relations by showing that the operator is protecting the off-site area. Public officials are insistent that the environmental monitoring program shall show whether the facility is operating in a manner consistent with public health requirements.

A preoperational surveillance program might be considered unnecessary, since postoperational monitoring would be adequate to show whether plant discharges have resulted in levels above established maximum permissible concentrations. There are two reasons why this concept is not valid. First, maximum permissible concentrations have been established only for ingestion and inhalation, and it is quite possible that a discharge, acceptable on this basis, might result in unacceptably high concentrations in some portion of the environment due to biological uptake. Work at Hanford (3) has shown that this would be true for the P^{32} content of food fishes if the water concentration were at the MPC. Secondly, regulatory agencies may be expected to take an ultra-conservative position on the addition of appreciable quantities of radioactivity to the environment even where the levels remain well below maximum permissible concentrations. Therefore, the operator of any nuclear facility must be prepared to show that plant wastes have not substantially increased the environmental levels. For this the preoperational levels must be known.

Unfortunately these preoperational levels -- the "baseline" -- are not like the everlasting hills of the prophet. They are more like a mirage, changing from place to place and from time to time. The "baseline" activity is contributed by a number of sources, each of which varies from time to time in

amount and nature. However, each of these sources is limited in isotopic composition, and radionuclide analysis will tend to show how each source is contributing to the total activity.

Every site will have some activity due to natural sources. The air will contain radon and its daughters, and probably some thoron, with its descendants. The biological environment will contain measurable activity, due to K^{40} and C^{14}. Most soils will contain some uranium, and thorium is even more common. In many cases, additional natural radioactivity will be found, for example, radium in the ground water in Northern Illinois (11). The levels of natural activity will change from time to time because of weather, changes in flow of streams or ground water, and the like.

Another ubiquitous source of radioactivity is weapons test fallout, consisting of a large variety of radionuclides. In addition to at least thirty important fission products, such induced activities as hydrogen-3 (2), carbon-14 (9), zinc-65 (4, 5, 8, 13), iron-55 and -59 (1,8,13), cobalt-57, -58, and -60 (8, 12, 13), manganese-54 (10), scandium-46 (7), cesium-134 (7), uranium-237 (6), calcium-45 (6), and probably still others have been identified. With such a variety of nuclides, all decaying with different half-lives and all having been distributed in a manner dependent on meteorological conditions and test operations, it is obvious that the fission product baseline will vary widely from place to place and from time to time.

While natural activity and fallout are probably the largest contributors to preoperational radioactivity at the present time, the possibility of contamination by users of "byproduct material" or from other nuclear facilities must not be neglected. As nuclear operations and the use of radioactivity become more common, this source of activity can be expected to become more important.

A suitable preoperational radionuclide assay program, then, can establish the sources of the "background" activity and can explain changes in its level. But this is not the only advantage to be obtained from preoperational nuclide analysis. An operator can, by selecting those nuclides with which he is primarily concerned, establish a baseline for those nuclides which is far more meaningful than data on gross levels of activity.

A preoperational monitoring program must cover the same types of samples to be used in the postoperational environmental surveillance. These, in turn, will depend on the nature of the planned operation and on the environment of the site. If the plant discharges are predominantly liquid, attention should be focussed on streams and ground water, and on the biota which they support. If wastes are largely airborne, the major part of the surveillance program should be devoted to air particulates and to vegetation in the path of the prevailing winds. Environmental (including economic) factors will also play a part: more attention will be paid to areas of high population densities, and

foodstuffs -- crops, shellfish, fish, milk -- will be chosen in preference to other biological samples.

Once the preoperational sampling program has been decided, on the basis of the criteria in the preceding paragraph, it is possible to decide which radionuclide analyses will be most useful. Three factors must be considered here -- the importance of the nuclides in plant operation, the relative hazard, and the tendency to concentrate in the samples (particularly the biological samples). Because of the expense of radionuclide analyses, the bulk of the monitoring program will always be dependent on gross analyses. Only a fraction of the samples -- usually the more active ones -- will be analyzed isotopically. Even in these, complete isotopic analysis will be neither feasible nor necessary. The most useful tool for this analysis is probably multi-channel gamma spectroscopy. This has the advantages of determining a number of nuclides simultaneously, of requiring minimal sample preparation, and of giving little or no opportunity for contamination or chemical loss. The disadvantage of low sensitivity, as compared to radiochemistry followed by beta counting, is partially overcome by its ready adaptability to very large samples. Where gamma spectroscopy is not appropriate, one must turn to radiochemical techniques or to more specialized instrumentation techniques, such as liquid scintillation for tritium. In some cases, biochemistry may be substituted

for radiochemistry, as in the use of rabbit thyroids in environmental monitoring for radioiodine.

Let us now consider some of the nuclides which may well be determined at different types of installations. As one such facility, consider a plant located in a rural area for the reprocessing of reactor fuel elements. Here the major wastes are gases freed in the dissolution step, and liquid process wastes. The nuclides of major concern would be the longer-lived fission products, particularly strontium-90 and cesium-137, but also including a number of rare-earth elements, zirconium-niobium, and ruthenium. Next in importance would come the alpha emitters, plutonium and uranium. The types of samples to be emphasized are surface and ground waters, since the most active waste is liquid, and also soil and agricultural products. The insoluble materials, plutonium, rare earth elements, zirconium-niobium, and ruthenium would be expected to collect on silts and soil, while the more soluble materials, such as strontium and cesium, would be expected to be more prominent in the water and crops. A suitable sampling and analysis program can then be established, and the preoperational levels of these materials in the environment determined over a considerable span of time. With such information, the plant operator has as well-defined a baseline as he can hope for, and the contributions (or hopefully the lack thereof) of his plant to the environmental radioactivity load can be evaluated fairly.

As a second example, consider a power reactor of the PWR type located slightly upstream of an estuary in which there are commercial shellfish and fishing. The design calls for bleeding a small part of the primary coolant continuously to waste. In this case, the major radionuclides are activated corrosion products, plus fission products which leak through imperfections in the cladding. The chief environmental factors are the water, and especially the commercial shellfish and fish. The shellfish, in particular, have been reported to concentrate radio-iron, -cobalt, and -zinc to a very large extent. Emphasis on these nuclides in preoperational monitoring will make it possible to determine whether a later rise in activity can be fairly attributed to the plant. Certain indicator fission products, chosen for their high yield and for ease of analysis, may be used to check for fission product leakage. Typical of such nuclides are iodine-131 and barium-140, probably best found in the algae of the stream. Again, determination of the levels of these nuclides before operation would be of value in deciding whether the plant had significantly raised the environmental values.

For a third example, consider a plant utilizing a few different radioactive materials. To be specific, this plant might be making tritium-activated self-luminous phosphors, cesium-137 and europium gamma sources, and strontium-90 medical applicators. Obviously this plant will contribute only a very

limited spectrum of radioactivity to the environment, and its preoperational monitoring should be directed toward the determination of these isotopes, particularly in any part of the environment in which they may be expected to concentrate. If such an organization should decide to make a new product, it would be desirable to start a new monitoring program, preoperational as far as that material is concerned, before entering into production.

September 17, 1958

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